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For: THERMOPLASTIC SHAPED-BODY...	)	

REQUEST FOR PRIORITY

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

In accordance with the provisions of 37 C.F.R. §1.55 and the requirements of 35 U.S.C. §119, there is filed herewith a certified copy of: **JAPANESE**

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It is respectfully requested that applicant be granted the benefit of the priority date of the foreign application.

Respectfully submitted,

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【書類名】　明細書

【発明の名称】　熱可塑性トレハロース成形物とその製造方法並びに用途

【特許請求の範囲】

【請求項１】　熱可塑性トレハロース成形物。

【請求項２】　水分を、約２．４ｗ／ｗ％以下含有している請求項１記載の熱可塑性トレハロース成形物。

【請求項３】　流動可能な温度が、品温として約９０乃至１２０℃又はこれ以上の温度である請求項１又は２記載の熱可塑性トレハロース成形物。

【請求項４】　トレハロースが非晶質である請求項１、２又は３記載の熱可塑性トレハロース成形物。

【請求項５】　成形物が、短径として約０．１乃至２０ｍｍから選ばれる長さを有し、長径として短径と同等以上の長さを有する請求項１、２、３又は４記載の熱可塑性トレハロース成形物。

【請求項６】　成形物が、粒、球、半球、楕円球、短棒、短角錘、半円錘、短角錐、立方体、直方体、短管及び繊維から選ばれる形状である請求項１、２、３、４又は５記載の熱可塑性トレハロース成形物。

【請求項７】　水分を、約２．４ｗ／ｗ％以下含有しているトレハロース溶液を成形することを特徴とする熱可塑性トレハロース成形物の製造方法。

【請求項８】　トレハロース溶液が、トレハロース含水結晶又はトレハロース無水結晶を加熱溶融したものであるか、又はトレハロース溶液を加熱濃縮したものである請求項７記載の熱可塑性トレハロース成形物の製造方法。

【請求項９】　トレハロース溶液が、無水物換算で、トレハロースを、６０ｗ／ｗ％を越える量含有している請求項７又は８記載の熱可塑性トレハロース成形物の製造方法。

【請求項１０】　トレハロース溶液を加熱濃縮するに際し、付着力低減能を有する物質と共存せしめることを特徴とする請求項９記載の熱可塑性トレハロース成形物の製造方法。

【請求項１１】　請求項１乃至６のいずれかに記載の熱可塑性トレハロース成形物、又は請求項７乃至１０のいずれかに記載の製造方法で得られた熱可塑性

トレハロース成形物を加熱して流動可能とし、これをそのまま成形するか、又は、これを他の物質と共に用いて成形することを特徴とするトレハロース含有成形物の製造方法。

【請求項１２】　熱可塑性トレハロース成形物の流動可能な温度が、品温として約９０乃至１２０℃又はこれ以上の温度である請求項１１記載のトレハロース含有成形物の製造方法。

【請求項１３】　熱可塑性トレハロース成形物を他の物質に対して、無水物換算で１．０ｗ／ｗ％以上の量用いることを特徴とする請求項１１又は１２記載のトレハロース含有成形物の製造方法。

【請求項１４】　他の物質が、飲食品材料、化粧品材料、医薬品材料、農水畜産材料及びプラスチック材料から選ばれる材料である請求項１１、１２又は１３記載のトレハロース含有成形物の製造方法。

【請求項１５】　成形する方法が、スタンピング成形、カッティング成形、晶出成形、射出成形、押出成形、吹込成形、板成形、注型成形、加圧成形、圧縮成形、カレンダ成形、積層成形、被覆成形、回転成形、吹付成形、固着成形及び発泡成形から選ばれる請求項１１、１２、１３又は１４記載のトレハロース含有成形物の製造方法。

【請求項１６】　トレハロース含有成形物が、飲食品、化粧品、医薬品、農水畜産用品、除崩性プラスチック又は家庭用品として利用される成形物である請求項１０、１１、１２、１３、１４又は１５記載のトレハロース含有成形物の製造方法。

【請求項１７】　トレハロース溶液を、加熱濃縮するに際し、付着力低減能を有する物質と共存せしめるか、又は共存せしめないままで水分を２．４ｗ／ｗ％以下にせしめることを特徴とするトレハロース溶液の付着力低減方法。

【請求項１８】　付着力低減能を有する物質が疎水性又は親水性の有機物質である請求項１７記載の付着力低減方法。

【請求項１９】　有機物質が、脂質、炭化水素、糖質、ポリアルコールから選ばれる請求項１７又は１８記載の付着力低減方法。

【請求項２０】　付着力低減能を有する物質を、無水物換算で、トレハロー

スに対して４０ｗ／ｗ％未満の量共存せしめることを特徴とする請求項１７、１８又は１９記載の付着力低減方法。

【請求項２１】　請求項１７乃至２０のいずれかに記載の付着力低減方法で付着力を低減させた、水分２．４ｗ／ｗ％以下のトレハロース溶液を成形し、得られる熱可塑性トレハロース成形物。

【請求項２２】　流動可能温度が、品温として約９０乃至１２０℃又はこれ以上の温度である請求項２１記載の熱可塑性トレハロース成形物。

【発明の詳細な説明】

【０００１】

【発明の属する技術分野】

本発明は、熱可塑性トレハロース成形物とその製造方法並びに用途に関し、詳細には、熱可塑性トレハロース成形物とこれをトレハロース溶液を用いて成形することを特徴とする当該熱可塑性トレハロース成形物の製造方法並びに当該成形物を加熱して流動可能とし、成形することを特徴とするトレハロース含有成形物の製造方法、加えてトレハロース溶液の付着力低減方法とその用途に関する。

【０００２】

【従来の技術】

トレハロースは、「化学工業」１９９９年６月号５７乃至６５頁の記載にも見られるように、近年、澱粉を原料に大量に生産され、食品工業を中心に広く利用されるようになってきた。しかしながら、トレハロースを成形して利用する方法は限られている。例えば、結晶又は粉末状のトレハロースを賦形剤又は結合剤などと混合し、錠剤に成形して利用するか、又は結晶又は粉末状のトレハロースをこれを越える量の他の糖質と共に水に加熱溶解し、これを煮詰めて型に流し込み、冷却して、キャンディーに成形して利用する程度である。しかも、これらの方法は、比較的特殊な装置、高度な技術を必要としているにもかかわらず、得られる成形物の形状が単調に過ぎる。随意な形状のトレハロース含有成形物、例えば綿状成形物の製造は困難で、市販の結晶又は粉末状トレハロースは、綿菓子製造機にかからず、氷砂糖を用いた場合のようには上手く綿状成形物が製造できないのが実状である。トレハロースを用いて、随意な形状の成形物を容易に製造する方

法が望まれる。

【０００３】

【発明が解決しようとする課題】

本発明は、トレハロースを用いて、随意な形状の成形物を容易に製造することを課題とする。

【０００４】

【課題を解決するための手段】

本発明者は、上記課題を解決するために、トレハロース成形物の製造方法につき、鋭意研究してきた。その結果、トレハロース高含有のガラス質成形物は、他の糖質の場合よりも保存安定性が良好であること、熱可塑性を有していること、耐熱性が良好であること、加熱して流動可能にしたものを随意な形状に成形することの容易であること、更には、これら性質を利用して繰り返し成形することも容易であることを見出し、本発明を完成した。即ち、本発明は、トレハロース高含有のガラス質成形物が優れた熱可塑性を有し、安定な成形物であることを見いだしたことに基づき、熱可塑性トレハロース成形物を提供し、又、水分が、２．４ｗ／ｗ％（以下、本明細書では、特に断らない限り、ｗ／ｗ％を％と略記する。）以下のトレハロース溶液を用いて成形することを特徴とする当該熱可塑性トレハロース成形物の製造方法を提供し、又、当該熱可塑性トレハロース成形物を加熱して流動可能とし、そのまま随意の形状に成形するか、又はこれを他の物質と共に用いて随意の形状に成形することを特徴とするトレハロース含有成形物の製造方法を提供し、更に、トレハロース溶液を加熱濃縮するに際し、付着力低減能を有する物質を共存せしめるか、又は共存せしめないままで水分を２．４％以下にせしめることを特徴とするトレハロース溶液の付着力低減方法とこの方法を利用し得られる当該熱可塑性トレハロース成形物を確立して前記課題を解決する。

【０００５】

【発明の実施の形態】

本発明で使用するトレハロースは、別名α、α－トレハロースであり、その由来は問わない。例えば、特開平７－２４６０９７号公報に記載される酵母からの

トレハロース、特開昭５８－２１６６９５号公報に記載されるマルトースからのホスホリラーゼ法によるトレハロース、特開平７－１７０９７７号公報、特開平７－２１３２８３号公報等に記載される澱粉からの酵素糖化法によるトレハロース等各種トレハロースが適宜採用できる。市販の高純度トレハロース含水結晶、高純度トレハロース無水結晶等を使用することも適宜採用できる。例えば、株式会社林原商事が販売している高純度トレハロース含水結晶製品（登録商標『トレハオース』）を使用することも有利に実施できる。

【０００６】

また、本発明の熱可塑性トレハロース成形物は、室内の温度、湿度環境条件下で保存安定性が良好で、経日変化を起こしにくく、しかも、これを加熱して溶融させ流動可能とし、随意の形状に容易に成形でき、成形、溶融を二度以上繰り返してもその性質に変化がなく、成形物の再生も容易に行えるものである。

【０００７】

本発明の熱可塑性トレハロース成形物は、水分約２．４％以下、望ましくは２．０％以下を有し、品温を約７０乃至９０℃にすると柔らかくなり、約９０乃至１２０℃又はこれ以上の温度にすると流動可能となる成形物である。その製造方法としては、適宜の方法が選択できる。例えば、トレハロースの含水結晶又は無水結晶を、そのまま又は適量の加湿をして、加熱、溶融、望ましくは品温としてできるだけ高温、好ましくは、１６０℃を越える温度、更に望ましくは約１６５乃至２３０℃、更に望ましくは、約１７０乃至２２０℃で加熱溶融させ、これをガラス質状態に成形する方法で成形すればよい。例えば、前記の各種酵素反応により調製したトレハロース溶液や、これを水素添加して調製したトレハロース溶液、更にはトレハロース含水結晶を水に溶解したトレハロース溶液、望ましくは水分３０％以上、換算すれば、トレハロースを無水物換算で７０％未満、更に望ましくは約５０％以上７０％未満含有するトレハロース溶液を、加熱濃縮して水分約２．４％以下、望ましくは約２．０％以下の高濃度溶液とし、これをガラス質状態に成形する方法で製造すればよい。

【０００８】

このトレハロース溶液を加熱濃縮する場合には、その水分と溶液温度とは、密

接に関係しており、例えば、常圧の場合、水分１５．３％で温度１１５℃であり、以下、同様に９．９％で１２５℃、５．２％で１４５℃、２．４％で１６５℃、１．１％で１８５℃の関係にある。この関係を求める過程で、加熱濃縮時の溶液の水分（溶液の温度）の違いによって溶液の付着力が大きく変動するという意外な現象を見出した。即ち、加熱濃縮して、溶液の水分が約１０％を下回ると急激に付着力が高まり、更に濃縮して水分約４乃至３％で付着力はピークに達し流動性が弱まり、更に加熱濃縮を続け、水分３％未満になると付着力が急激に低下し、更に水分約２．４％以下、望ましくは約２．０％以下になるとその付着力、流動性も成形時の作業性に支障がない程に改善されることが判明した。

【０００９】

この異常に高まった付着力を低減させ作業性を改善することを目的に鋭意検討したところ、付着力低減能を有する物質の共存が好適であり、これにより、高純度トレハロース溶液の付着力が大幅に低減され、加熱濃縮時の泡立ちが低くなり、作業性が向上し、成形物の製造も容易となることが判明した。

【００１０】

付着力低減能を有する物質としては、前記トレハロース溶液の付着力が低減できる物質であればよく、望ましくは、脂質、炭化水素などの疎水性物質や、糖質、ポリアルコールなどの親水性物質などから選ばれる有機物質が好適である。

【００１１】

脂質としては、油脂、脂肪酸など、炭化水素としては、約１００乃至２３０℃で液状を示す、例えば、アルカン、アルケン、複素環化合物など、糖質としては、単糖類、オリゴ糖、単糖アルコール、二糖アルコールなど、ポリアルコールとしては、グリセリン、ジエチレングリコール、プロピレングリコールなどから１種又は２種以上のものが適宜選ばれる。

【００１２】

以上述べてきたように、トレハロース溶液は、望ましくは、付着力低減方法を採用して、所定の水分、望ましくは２．４％以下に濃縮し、これをガラス質状態に成形して当該熱可塑性トレハロース成形物を製造すればよい。成形方法としては、公知の各種の方法、例えば、スタンピング成形、カッティング成形、大気中

成形、水中又は油中成形、押出成形、加圧成形、とりわけ二軸式押出成形などが有利に実施できる。

【００１３】

このようにして得られる熱可塑性トレハロース成形物は、その良好な保存安定性、熱可塑性を保つ上で、できるだけトレハロース含量の高いものが望ましく、通常、無水物換算でトレハロースを６０％を越える量含有するのが好適であり、望ましくは約７０％以上、更に望ましくは約８０％以上、更に望ましくは９０％以上含有しているものが好適である。

【００１４】

熱可塑性トレハロース成形物の形状としては、通常、これを原料とし、加熱して流動可能とし随意の形状に成形する際に、公知の各種成形機又は成形装置に容易に適用できるものが望ましく、これら成形機又は成形装置を利用して、容易に加熱し、必要に応じて加圧又は減圧して、容易に溶融させ流動可能とし、随意の形状に成形できるものが好適である。

【００１５】

この具体例としては、通常、短径として約０．１乃至２０ｍｍ、望ましくは約２乃至１５ｍｍから選ばれる長さを有し、長径として短径と同等以上の長さを有するものが好適である。形状としては、粒、球、半球、楕円球、短棒、短角錐、半円錐、短角錐、立方体、直方体、短管、繊維などが有利に利用できる。一般的には、ペレットと称される形状が好適である。

【００１６】

このようにして得られた熱可塑性トレハロース成形物は、通常、水分約２．４％以下、望ましくは約２．０％以下、トレハロースを無水物換算で６０％を越える量、望ましくは約７０％以上を含有するトレハロース成形物であって、しかも、トレハロースが非晶質で透明乃至半透明で、保存安定性、耐熱性に優れ、取扱い容易である。当該熱可塑性トレハロース成形物の用途としては、例えば、これを少量の熱水に溶解して、トレハロース高含有ペースト状物を調製することや、更にこのペースト状物を他の機能性物質や有効成分と混合して用いることや、更にはこの混合物にトレハロースの結晶核などを共存させ、トレハロースを起晶さ

せて粉末化を容易にする粉末化基剤などとして用いたりすることもできる。

【００１７】

しかし、最も重要な用途は、熱可塑性を利用する用途である。即ち、熱可塑性トレハロース成形物は、これを再び加熱して溶融させ流動可能とし、これをそのまま随意の形状に成形してもよいし、又、これを他の物質と共に用いて、例えば、これを他の物質に被覆、混合、固着、挿入、封入させるなどして、随意の形状に成形してトレハロース含有成形物を得ることも有利に実施できる。

【００１８】

加熱する条件としては、熱可塑性トレハロース成形物が溶融して柔らかくなり、望ましくは流れる状態、換言すれば、流動可能となり、随意の形状に成形できればよく、柔らかくなる温度としては、品温約７０乃至９０℃、流動可能温度としては、品温約９０乃至１２０℃又はこれ以上に加熱すればよい。具体的には、流動可能温度は、通常、当該熱可塑性トレハロース成形物の水分が比較的高い約２．４％未満１．５％以上の場合、比較的低く、通常、約９０℃以上、望ましくは約１００乃至１６０℃から選ばれ、その水分が比較的低い約１．５％未満の場合、比較的高く、通常、約１００℃以上、望ましくは約１１０乃至２３０℃から選ばれる温度に加熱すればよい。

【００１９】

他の物質と共に用いて成形する際には、例えば、当該熱可塑性トレハロース成形物を、予め他の物質と混合し、次いで、加熱して、流動可能として随意の形状に成形することも、又、当該熱可塑性トレハロース成形物だけを予め加熱して流動可能とし、次いで他の物質と混合して成形することも適宜選択できる。必要ならば、加湿するか、又は少量の水を加えて、溶融温度を下げたり、混和を促進したりすることもできる。

【００２０】

他の物質としては、共に用いて随意の安定な形状に成形できるものであればいずれも有利に用いることができる。即ち、化学工業分野をはじめとする本発明の成形物が利用される個々の分野、例えば、飲食品分野、化粧品分野、医薬品分野、農水畜産分野、プラスチック工業、家庭用品などの分野において通常用いられ

る添加物を含む材料ないしは素材であって、共に用いて安定な形状に成形できるものであればいずれでもよい。詳細には、上記の分野から選ばれる１又は２以上の分野で通常用いられる、例えば、甘味料、着色剤、発色剤、発酵調整剤、着香料、香料、芳香剤、強化剤、膨張剤、保存料、殺菌料、酸化防止剤、抗酸化剤、漂白剤、糊料、安定剤、乳化剤、軟化剤、品質改良剤、調味料、香辛料、光沢剤、苦味料、酸味料、乳化剤、食品製造用剤、酵素剤、ガムベース、界面活性剤、可塑剤、滑沢剤、可溶化剤、還元剤、緩衝剤、基材、吸着剤、矯味剤、結合剤、懸濁剤、コーティング剤、潤滑剤、潤滑調整剤、充填剤、消泡剤、清涼化剤、接着剤、増強剤、咀嚼剤、糖衣剤、等張化剤、粘着剤、粘着増強剤、粘稠剤、粘稠化剤、発炎抑制剤、発熱剤、発泡剤、皮膚保護剤、賦形剤、分散剤、ｐＨ調節剤、崩壊剤、崩壊補助剤、防錆剤、防湿剤、防腐剤、防黴剤、保存剤、無痛化剤、ビタミン剤、抗生物質、化学療法剤、アレルギー用剤、抗ヒスタミン剤、鎮痛剤、鎮痒剤、収斂剤、消炎剤、ステロイド剤、ホルモン剤、強心剤、歯科・口腔用剤、止血剤、紫外線吸収剤、除草剤、熱可塑性樹脂、合成樹脂、劣化防止剤、老化防止剤、焼け防止剤、樹脂硬化剤、凝固剤、起泡剤、気泡安定剤、帯電防止剤、加硫剤などから目的に応じて適宜選択することができる。例えば、飲食品分野でより具体的に述べれば、例えば、澱粉、アミロース、アミロペクチン、α化澱粉、デキストリン、難消化性デキストリン、食物繊維、オリゴ糖、機能性オリゴ糖、単糖、糖アルコールなどの糖質、蛋白質、ペプチド、アミノ酸、油脂、脂肪酸、ミネラルなどが挙げられる。

【００２１】

このようにして他の物質と共に用いて成形し得られるトレハロース含有成形物に含まれるトレハロースは、通常、前記ビタミン、脂質、蛋白質、薬効物質、生理活性物質などの比較的不安定な機能性物質や有効成分の安定性を向上させることから、その含量が高い程望ましく、一般的には、表面を被覆する成形物の場合に比較的少なく、均一に混合し全体を固着成形させるような場合には比較的多い。随意の形状に成形する必要があることから、通常、熱可塑性トレハロース成形物を他の物質に対して、無水物換算で、約１．０％以上、望ましくは、約２．０％以上、より望ましくは約５．０％以上、更に望ましくは約１０％以上が好適で

ある。成形物の形状としては、粒、棒、板、シート、フィルム、管、積層、スポンジなどの点、線、面、立体の各種形状の成形物が適宜選ばれる。

【００２２】

これら成形物を成形する方法は、目的とする形状によって、適宜の方法が採用できる。公知の成形方法、例えば、スタンピング成形、カッティング成形、射出成形、押出成形、吹込成形、板成形、注型成形、圧縮成形、加圧成形、カレンダ成形、積層成形、被膜成形、回転成形、吹付成形、固着成形、発泡成形などが有利に実施できる。

【００２３】

このようにして、他の形状に変換して得られるトレハロース含有成形物は、共に用いる他の物質やこれに含まれる機能性物質や有効成分などによって、例えば、飲食品、化粧品、医薬品、農水畜産用品、徐崩性プラスチック、家庭用品などとして利用される成形物に好適である。

【００２４】

以下、本発明を実験で説明する。

【００２５】

【実験１】

〈糖質溶液の煮詰温度（水分）による溶液の付着性とその成形物の保存安定性に与える影響〉

糖質として、非還元性の糖質、即ちマルチトール無水結晶又はトレハロース２含水結晶を用意し、手鍋にこれら糖質のいずれかをとり、これに水を加えて加熱し、それぞれ濃度５０％の溶液を調製し、これら糖質溶液の加熱を続け沸騰させ、糖質溶液を煮詰めた。糖質溶液が、１２０℃、１３０℃、１４０℃、１５０℃、１６０℃、１７０℃、１８０℃、１９０℃の各温度に達した時、糖質溶液にほぼ垂直に、直径４ｍｍのガラス棒を液面から３ｃｍの深さまで差込み、そのまま約３秒間保持した後、緩やかに引き上げた。ガラス棒に付着した糖質溶液の重量は、糖質溶液が付着したガラス棒の重量から、ガラス棒のみの重量を差し引くことによって、算出した。また、それぞれの煮詰め液を小型成形器（デポジッター）にサンプリングし、室温まで冷却させ、形状が、長さ２０ｍｍ、幅１５ｍｍ、

厚さ５ｍｍの成形物の試料を得た。この製造当日の試料を用いて、水分を測定した。水分含量は、常法により、珪藻土法で測定した。

【００２６】

また、同じ試料をシャーレに入れて、相対湿度約７０％で、温度約２５℃の室内に１０日間放置後の固着性の有無を観察した。

【００２７】

固着性は、試料が吸湿し、シャーレに固着する程度を調べ、有、無の２段階に分けて示し、保存安定性の目安とした。結果は、表１に示した。

【００２８】

【表１】

糖質	測定項目	煮詰温度(常圧，℃)									
		120	130	140	150	155	160	165	170	180	190
トレハロース	付着量(g)	0.230	0.569	0.890	1.250	1.474	1.726	1.580	0.915	0.761	0.579
	水分(%)	12.1	8.3	6.1	4.4	3.6	3.0	2.4	2.0	1.3	0.9
	固着性	無	無	無	無	無	無	無	無	無	無
マルチトール	付着量(g)	0.200	0.317	0.338	0.369	0.355	0.335	0.345	0.313	0.294	0.257
	水分(%)	9.1	5.5	4.1	3.2	2.7	2.5	2.1	1.9	1.4	0.9
	固着性	有	有	有	有	有	有	有	有	有	有

【００２９】

表１から明らかなように、トレハロースの場合、温度１２０℃（溶液の水分約１２％）から１６０℃（溶液の水分約３％）まで、ガラス棒に付着するトレハロース溶液の重量は著しく増加し、この温度を越えると、水分が減少するにもかかわらず、温度１９０℃まで付着量が急激に減少することがわかった。即ち、トレハロース溶液の付着力は、温度約１５０℃乃至１６０℃（溶液の水分約４乃至３％）にピークに達し、１６０℃を越える（溶液の水分約３％未満になる）とその付着力が漸次低下することが判明した。

【００３０】

なお、対照のマルチトールの場合、付着量の僅かな増加と減少（温度１２０℃から１５０℃までの増加と、その後の温度１９０℃までの減少）が観察されたが、その増減の程度は、トレハロースの場合と比べ、微少であることが判明した。以上のことから、トレハロース溶液を煮詰める際、温度１６０℃までは急激に付着力が上昇し、成形時の作業性に悪影響を及ぼすので、これを改善する必要のあることが判明した。一方、このトレハロース溶液を温度１６５℃以上（溶液の水分２．４％以下）の高温に煮詰めると付着力が低下し、流動性が著しく改善され、トレハロース溶液を取扱う際の作業性が極めて容易になることも判明した。トレハロースの場合はマルチトールとは違って、成形物は、相対湿度７０％の室温下に保存しても経時変化が少なく、安定性良好であることが判明した。

【００３１】

【実験２】

〈付着力の低減方法〉

トレハロース２含水結晶５５０重量部をステンレス製の手鍋に入れ、そこへ付着力低減能を有する物質として、サフラワー油５０重量部、サラダ油５０重量部、マルチトール無水結晶１５０重量部、又は、ラクチトール１含水結晶１５８重量部から選ばれるいずれか１種を加え、更に水４５０重量部を加えた後、加熱しながら糖質を完全に溶解させた。更に、加熱を続け沸騰させ、糖質溶液を煮詰めた。糖質溶液が、１２０℃、１３０℃、１４０℃、１５０℃、１６０℃、１７０℃、１８０℃の各温度に達した時、実験１と同様の方法で、ガラス棒に付着する糖質溶液の重量を測定した。結果は、表２に示した。なお、糖質溶液中のトレハロース含量は、無水物換算で算出し、表２に併せて示した。

【００３２】

【表２】

糖質	共存物質 トレハロース含量 (無水物換算)	測定項目	煮詰温度（常圧，℃）						
			120	130	140	150	160	170	180
トレハロース	無 100%	付着量 (g)	0.230	0.569	0.890	1.250	1.726	0.915	0.761
	サフラワー油 約91%	付着量 (g)	0.150	0.194	0.225	0.256	0.257	0.277	0.278
	サラダ油 約91%	付着量 (g)	0.195	0.499	0.512	0.596	0.538	0.481	0.428
	マルチトール 約77%	付着量 (g)	0.230	0.538	0.861	0.987	0.786	0.607	0.570
	ラクチトール 約77%	付着量 (g)	0.265	0.466	0.773	0.862	0.919	0.893	0.574

【００３３】

表２から明らかなように、トレハロース単独の場合の温度約１５０℃乃至１６０℃における急激な付着力の増加は、サフラワー油、サラダ油、マルチトール、ラクチトール共存下のいずれの場合も、顕著に抑制され、その付着力が激減することが判明した。

【００３４】

更に、温度１６０℃を越す高温での付着力も、これら付着力低減能を有する物質の存在下では、トレハロース単独の場合と比べ、その付着力はいずれも低いことが判明した。

【００３５】

以上のことから、トレハロース溶液を煮詰め中に認められた温度１６０℃までの急激な付着力の上昇は、サフラワー油、サラダ油などの油脂、マルチトール、ラクチトールなどの糖質など付着力低減能を有する物質を煮詰の際に共存させることにより、著しく抑制され、更に、温度１６０℃を越す高温煮詰においても、これら物質がトレハロース溶液の付着力を一層低下させ、流動性を著しく改善することがわかり、トレハロースを有効成分とする熱可塑性トレハロース成形物の成形時の作業性を顕著に改善できることが判明した。

【００３６】

以下、本発明の熱可塑性トレハロース成形物の製造方法とその用途例の２～３を説明する。

【００３７】

【実施例１　短棒状成形物】

高純度トレハロース含水結晶製品（株式会社林原商事販売、登録商標『トレハオース』）１１０重量部を溶解タンクにとり、水９０重量部を加えて加熱溶解し、これを手鍋にとって加熱濃縮し、溶液温度約１９０℃（水分約０．９％）まで煮詰めて溶液の付着力を低減させた後、これを押出成形機にかけて、成形し、室温まで放冷して直径約２ｍｍ、長さ約４ｍｍの短棒状の熱可塑性トレハロース成形物を得た。本品は、無色で透明乃至半透明な実質的に非晶質な成形物で、室内環境での保存安定性が良好で、熱可塑性を有していることから、これを約９０℃以上に再加熱して流動可能とし、随意な他の形状の成形物に変化させることも、又、これを各種機能を有する他の物質と共に用い、随意な形状の成形物に仕上げることも有利に実施できる。また本品は、比較的少量の熱水に溶解させてトレハロース含有ペーストを調製したり、更に、このペーストを非晶質のままで利用、又は結晶化させて用いる粉末化基剤として利用することも有利に実施できる。

【００３８】

【実施例２　楕円球状成形物】

高純度トレハロース含水結晶製品を実施例１と同様に加熱溶解し、これを手鍋にとって、加熱濃縮し、付着力が高まり始める液温が１１０℃を越える付近で、無水物換算で、トレハロースに対し約５％のオリーブ油を加え、溶液の付着力を低減させて加熱濃縮を続け、溶液温度約１７０℃（水分約２．０％）まで煮詰め、これをスタンピング成形機にかけて、長さ約１５ｍｍ、楕円球状の熱可塑性トレハロース成形物を得た。本品は、実施例１の成形物と同様に、無色で透明乃至半透明な実質的に非晶質な成形物で、室内環境での保存安定性が良好で、熱可塑性を有していることから、これを約９０℃以上に加熱して流動可能とし、随意な他の形状の成形物に変化させることも、又、これを各種機能を有する他の物質と共に用いて、随意な形状の成形物に仕上げることも有利に実施できる。また本品は、比較的少量の熱水に溶解させてトレハロース含有ペーストを調製したり、更

に、ペーストを非晶質のままで利用、又は結晶化させて用いる粉末化基剤として利用することも有利に実施できる。

【００３９】

【実施例３　短四角柱状成形物】

高純度トレハロース含水結晶製品を実施例１と同様に加熱溶解し、これを手鍋にとって加熱濃縮し、付着力が高まり始める液温が１１０℃を越える付近で、無水物換算でトレハロースに対して約３％のプロピレングリコールを加え、溶液の付着力を低減させて加熱濃縮を続け、溶液温度約１６５℃（水分約２．４％）まで煮詰め、これをカッティング成形機にかけて、一辺が約５ｍｍ、長さ約１０ｍｍの短四角柱状の熱可塑性トレハロース成形物を得た。本品は、実施例１の成形物と同様に、無色で透明乃至半透明な実質的に非晶質な成形物で、室内環境での保存安定性が良好で、熱可塑性を有していることから、これを約９０℃以上に再加熱して流動可能とし、随意な他の形状の成形物に変化させることも、又、これを各種機能を有する他の物質と共に用いて、随意な形状の成形物に仕上げることも有利に実施できる。また本品は、比較的少量の熱水に溶解させてトレハロース含有ペーストを調製したり、更に、このペーストを非晶質のままで利用、又は結晶化させて用いる粉末化基剤として利用することも有利に実施できる。

【００４０】

【実施例４　半球状成形物】

特開平８－７３４８２に開示される方法に従って、リゾビウム・スピーシーズＭ－１１（ＦＥＲＭ ＢＰ－４１３０）の変異株をファーメンターで栄養培地に約７０時間培養した。培養後、ＳＦ膜を用いて除菌濾過し、約１００１の培養濾液を回収し、更に、その濾液をＵＦ膜濃縮し、非還元性糖質生成酵素（約４１０単位／ｍｌ）とトレハロース遊離酵素（約４９０単位／ｍｌ）とを含む濃縮酵素液約５１を回収した。とうもろこし澱粉を濃度約３３％の澱粉乳とし、これにα－アミラーゼを作用させてＤＥ約４の液化溶液を得、次いで、前記方法で調製した非還元性糖質生成酵素とトレハロース遊離酵素とを含む濃縮酵素液を澱粉グラム当たり０．０２ｍｌ、イソアミラーゼを澱粉グラム当たり５００単位及びシクロマルトデキストリン・グルカノトランスフェラーゼを澱粉グラム当たり５単位

加え、ｐＨ６．２、温度４０℃で４８時間反応させた。本反応液を加熱失活し、次いで、グルコアミラーゼを基質グラム当たり１０単位加え、ｐＨ５．０、温度５０℃で１０時間反応させた。本反応液には、固形物当たりトレハロースを８５．６％含有していた。本反応液を加熱失活し、常法に従って、脱色、脱塩して精製し、濃縮して濃度４５％のシラップを得た。本トレハロース高含有低還元性糖質シラップを、常法に従って、水素添加し、脱色、脱塩して精製し、濃縮して、無水物換算でトレハロースを約８５％、残りは主としてソルビトールを含有するシラップを得た。これを手鍋にとって加熱濃縮し、溶液温度約１８０℃（水分約１．３％）まで煮詰めて溶液の付着力を低減させ、これを細管を通してサラダ油中に滴下して冷却固化させ、底面直径約８ｍｍの半球状の熱可塑性トレハロース成形物を得た。本品は、実施例１の成形物と同様に、無色で透明乃至半透明な実質的に非晶質な成形物で、室内環境での保存安定性が良好で、熱可塑性を有していることから、これを約９０℃以上に再加熱して流動可能とし、随意な形状の成形物に変化させることも、又、これを各種機能を有する他の物質と共に用いて、随意な形状の成形物に仕上げることも有利に実施できる。また本品は、比較的少量の熱水に溶解させてトレハロース含有ペーストを調製したり、更に、ペーストを非晶質のままで利用、又は結晶化させて用いる粉末化基剤として利用することも有利に実施できる。

【００４１】

【実施例５　繊維状成形物】

トレハロース含水結晶を電気炉で約２００乃至２１０℃（水分約０．５％以下）に加熱し、溶融させ、得られる溶液を、この電気炉の下部に設けた取り出し口から流出させつつ、これに空気を吹き付けて太さ約０．１乃至０．２ｍｍの繊維状物とし、これを更に破砕して長さ約２０ｍｍ以下の短繊維状物を得た。本品は、実施例１の成形物と同様に無色で透明乃至半透明な実質的に非晶質な成形物で、室内環境での保存安定性が良好で、熱可塑性を有していることから、これを約１００℃以上に再加熱して流動可能とし、随意な形状の成形物に変化させることも、又、これを各種機能を有する他の物質と共に用いて、随意な形状の成形物に仕上げることも有利に実施できる。また本品は、比較的少量の熱水に溶解させて

トレハロース含有ペーストを調製したり、更に、ペーストを非晶質のままで利用、又は結晶化させて用いる粉末化基剤として利用することも有利に実施できる。

【００４２】

【実施例６　短棒状成形物】

実施例３の方法で得た熱可塑性トレハロース成形物を、更に約１２０℃に加熱し押出成形機にかけて、直径約２ｍｍ、長さ約４ｍｍの短棒状の熱可塑性トレハロース成形物を得た。本品は、実施例１の成形物と同様に、無色で透明乃至半透明な実質的に非晶質な成形物で、室内環境での保存安定性が良好で、熱可塑性を有していることから、これを約９０℃以上に再加熱して流動可能とし、随意な形状の成形物に変化させることも、又、これを各種機能を有する他の物質と共に用いて、随意な形状の成形物に仕上げることも有利に実施できる。また本品は、比較的少量の熱水に溶解させてトレハロース含有ペーストを調製したり、更に、このペーストを非晶質のままで利用、又は結晶化させて用いる粉末化基剤として利用することも有利に実施できる。

【００４３】

【実施例７　綿状成形物】

実施例２の方法で得た熱可塑性トレハロース成形物１重量部に、レモン濃縮果汁０．０５重量部を均一に噴霧して付着させ、乾燥した後、これを綿菓子製造機にかけて約１００℃以上に加熱溶融し綿菓子を製造した。本品は、従来の砂糖を用いた綿菓子とは違って、酸味の効いたレモン風味のトレハロース含有綿菓子である。

【００４４】

【実施例８　被覆成形物】

小麦粉１００重量部、イースト２重量部、砂糖５重量部、マルトース１重量部及び無機フード０．１重量部を、常法に従って、水でこね、中種を２６℃で２時間発酵させ、その後、３０分間熟成し、得られたパン生地を手で丸めた。この表面に実施例４の方法で得た熱可塑性トレハロース成形物をパン生地に対し、無水物換算で、約２％を部分的に押し込んだ後、常法に従って約１８０℃で焼き上げた。本品は、表面に溶融変形した趣きのあるトレハロース含有被覆成形物を配置

したユニークなパンであり、又色相、すだちともに良好で適度な弾力、温和な甘味を有する高品質のパンである。

【００４５】

【実施例９　被覆成形物】

バター６０重量部、砂糖２０重量部、マルトース２０重量部及びトレハロース２含水結晶の粉末１０重量部をよく練り合わせ、これに鶏卵４０重量部を加えてクリーム状にする。更に、薄力粉１４０重量部、コーンスターチ１０重量部及びプルラン３重量部を加えて混合し、得られたクッキー生地を天板上に菊口金で絞り出して花模様に成形し、この上に、実施例５の方法で得た熱可塑性トレハロース成形物をクッキー生地に対し、無水物換算で約１％付着させて、オーブンにより約１７０℃で１５分間焼き上げた。この製品は、溶融変形した趣きのあるトレハロース含有被覆成形物とクッキー生地の肌別れもなく、充分接着しており、又、風味、口当たりも良好であった。

【００４６】

【実施例１０　顆粒状成形物】

実施例２の方法で得た熱可塑性トレハロース成形物１５重量部及びプレーンヨーグルト２重量部と無水トレハロース粉末１０重量部との混合粉末を、約９０℃に加熱し二軸式押出成形機にかけてトレハロース含有顆粒状成形物を製造した。本品は、ヨーグルト風味の甘味成形物で、プレミックス、冷菓、ケーキなどの製菓材料、経管流動食などの治療用栄養剤として、更には、例えば、マーガリンホイップクリーム、スプレッド、チーズケーキ、ゼリーなどに含有せしめヨーグルト風味の製品にするなど有利に利用できる。

【００４７】

【実施例１１　台形状成形物】

実施例３の方法で得た熱可塑性トレハロース成形物３０重量部、α－グルコシル－Ｌ－アスコルビン酸１重量部、α－グルコシルヘスペリジン１重量部、硫酸ナトリウム１重量部及び適量の着色料、香料を溶解したアルコール水１重量部を、約１００℃に加熱し加圧成形機にかけてトレハロース含有台形状成形物を製造した。本品は、１個約１０ｇで、美肌剤、色白剤として浴用に好適であり、浴槽

に沈めて利用すればよい。本品は、入浴用の場合と同様に、洗顔用水、化粧水などに溶解して利用することも有利に実施できる。

【００４８】

【実施例１２ 楕円球状成形物】

実施例２の方法で得た熱可塑性トレハロース成形物５００重量部、粉末卵黄２７０重量部、脱脂粉乳２０９重量部、塩化ナトリウム４．４重量部、塩化カリウム１．８５重量部、硫酸マグネシウム４重量部、チアミン０．０１重量部、アスコルビン酸ナトリウム０．１重量部、ビタミンＥアセテート０．６重量部及びニコチン酸アミド０．０４重量部の混合物を、約１００℃に加熱し加圧成形機にかけてトレハロース含有楕円球状成形物を製造した。本品は、栄養剤成形物として好適であり、１球約５ｇで、口で溶かして利用され、病人の治療用栄養剤として、又、健常人の健康の維持、増進などに有利に利用される。

【００４９】

【実施例１３ 顆粒状成形物】

実施例１の方法で得た熱可塑性トレハロース成形物１０重量部及び炭酸ナトリウム粉末１重量部の混合物を、約１００℃に加熱し二軸式押出成形機にかけてトレハロース含有顆粒状成形物を製造した。本品は、炭酸ナトリウム粉末の飛散の恐れのない取扱い容易なｐＨ調節剤として、各種用途に幅広く利用できる。

【００５０】

【実施例１４ 短管状成形物】

実施例２の方法で得た熱可塑性トレハロース成形物１００重量部、魚粉２００重量部、α化澱粉２０重量部、プルラン３重量部及び水１０重量部の混合物を、約１１０℃に加熱し押出成形機にかけて短管状成形物とし、これを約１０％セラックアルコール溶液に浸漬し、直ちに引き上げて熱風で乾燥して、トレハロース含有短管状成形物を製造した。本品は、水を汚染することの少ない飼料で、淡水、海水を問わず、養殖用、鑑賞用、魚介類、甲殻類などの飼料として好適である。

【００５１】

【実施例１５ 植木鉢】

実施例６の方法で得た熱可塑性トレハロース成形物１００重量部とグリセリン１５重量部及び古紙パルプ５０重量部の混合物を、約１５０℃に加熱し射出成形機にかけて植木鉢を成形し、溶融ワックスに浸漬し、放冷してトレハロース含有植木鉢を製造した。本品は、徐崩性、生分解性であるため、移植用植木鉢として好適であり、鉢をはずすことなく移植することができるので、植え痛みがない。

【００５２】

【実施例１６　短棒状成形物】

実施例１の方法で得た熱可塑性トレハロース成形物６０重量部、サンゴ粉末３０重量部及びコーンスティープリカー２重量部の混合物を、約１１０℃に加熱し押出成形機にかけて短棒状のトレハロース含有植物活性化剤成形物を得た。本品は、容器が不要で、取り扱いが容易であり、使用時に適度な速度で溶解、崩壊するので持続性植物活性化剤として好適である。

【００５３】

【実施例１７　ゴルフティー】

実施例４の方法で得た熱可塑性トレハロース成形物１０重量部、酸性白土４重量部及びプルラン０．５重量部の混合物を、約１６０℃に加熱し射出成形機にかけてゴルフティーを成形し、約１０％セラックアルコール溶液に浸し、乾燥して製品を得た。本品は、ショットにより、小塊に破壊され、雨水で徐々に崩壊し、生分解される。従って、ゴルフ場の美観を損なわず、環境も破壊しない。また、適度なトレハロースの散布は、ゴルフ場の芝の生育を活性化する。

【００５４】

【実施例１８　フィルム状成形物】

低密度ポリエチレン９２重量部、実施例３の方法で得た熱可塑性トレハロース成形物８重量部、ステアリン酸カルシウム０．０５重量部、酵素処理ルチン０．５重量部及び茶抽出部０．５重量部の混合物を、約１２０℃に加熱し加圧成形機にかけて短棒状成形物とし、次いで、これを常法に従って、約１３０℃に加熱し押出成形機（インフレーション法）にかけてフィルム状成形物を製造した。本品は、花、野菜、果物、肉、魚など生鮮食品の鮮度保持用フィルムとして有利に利用できる。

【００５５】

【発明の効果】

上記したように、本発明の熱可塑性トレハロース成形物は、良好な保存安定性、熱可塑性、耐熱性を有しており、この熱可塑性を利用して繰り返し成形できる特徴を有している。

【００５６】

この熱可塑性トレハロース成形物の製造に際し、トレハロース溶液の付着力を低減させて、成形時の作業性を改善することも有利に実施できる。

【００５７】

本発明の熱可塑性トレハロース成形物を加熱して流動可能とし、随意な形状の成形物を容易に製造することができる。この成形に際して、各種の機能性物質や有効成分、例えば飲食品材料、化粧品材料、医薬品材料、農水畜産材料、プラスチック成形材料などを混合して成形することも好都合である。

【００５８】

このようにして得られる成形物は、随意な形状に容易に成形できるだけでなく、トレハロースが比較的不安定な機能性物質や有効成分を安定化することから付加価値が向上し、加えて、トレハロースを含んでいることから、徐崩性、生分解性の成形物となり、地球環境に易しい成形物と言える。

【００５９】

従って、本発明が与える影響は、広範囲に及び、食品業界、化粧品業界、医薬品業界、農水畜産業界、プラスチック業界、家庭用品業界などの産業界に与える工業的意義は極めて大きい。

【書類名】　要約書

【要約】

【課題】　本発明は、トレハロースを用いて、随意な形状の成形物を容易に製造することを課題とする。

【解決手段】　熱可塑性トレハロース成形物を提供し、又、トレハロース溶液を用いて成形することを特徴とする当該熱可塑性トレハロース成形物の製造方法を提供し、併せて、当該熱可塑性トレハロース成形物を加熱して流動可能とし、随意な形状に成形することを特徴とするトレハロース含有成形物の製造方法を提供し、更に、トレハロース水溶液の付着力低減方法とその用途を提供して課題を解決する。

【選択図】　なし

認定・付加情報

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DECLARATION

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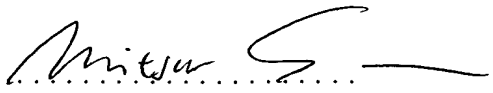
do solemnly and sincerely declare that I have a competent knowledge of English and Japanese languages and that the following is a true and accurate translation of the attached certificate numbered 2000-3089683 and dated 27th October 2000.

4th November 2000



Mitsuo SUMA

PATENT OFFICE

JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following application as filed with this Office.

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Application Number: Patent Application No. 299575/1999

Applicant(s): KABUSHIKI KAISHA HAYASHIBARA SEIBUTSU KAGAKU KENKYUJO

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[Document name] Specification

[Title of the Invention] Thermoplastic shaped-body of trehalose, process and uses thereof

[Claims] 1. A thermoplastic shaped-body of trehalose.

2. The thermoplastic shaped-body of trehalose of claim 1, which has a moisture content of not higher than about 2.4 w/w %.

3. The thermoplastic shaped-body of trehalose of claim 1 or 2, which has a free-flowing ability at temperatures of about 90°C to about 120°C or higher as a temperature thereof.

4. The thermoplastic shaped-body of trehalose of claim 1, 2 or 3, wherein said trehalose is amorphous.

5. The thermoplastic shaped-body of trehalose of any one of claims 1 to 4, which has a short diameter of about 0.1-20 mm and a long diameter of not shorter than the short diameter.

6. The thermoplastic shaped-body of trehalose of any one of claims 1 to 5, which has a form selected from the group consisting of granules, spheres, hemispheres, elliptical spheres, short rods, short pyramids, half corns, cubes, rectangular parallelepipeds, short tubes, and fibers.

7. A process for producing a thermostable shaped-body of trehalose, which comprises a step of forming a trehalose solution having a moisture content of not higher than about 2.4 w/w %.

8. The process of claim 7, wherein said trehalose solution is prepared by melting hydrous crystalline trehalose or anhydrous crystalline trehalose under heating conditions; or by concentrating a trehalose solution under heating conditions.

9. The process of claim 7 or 8, wherein said

thermoplastic shaped-body of trehalose contains trehalose in an amount of over 60 w/w %, on a dry solid basis.

10. The process of claim 9, wherein a substance capable of lowering the adhesion of a trehalose solution is coexisted during the concentration by heating of said trehalose solution.

11. A process for producing a shaped-body containing trehalose, which comprises the steps of heating either said thermoplastic shaped-body of trehalose of any one of claims 1 to 6 or said thermoplastic shaped-body of trehalose produced by the process of any one of claims 7 to 10 to impart a free-flowing ability thereunto, and forming the resultant alone or in combination with other substance.

12. The process of claim 11, wherein the temperature at which said thermoplastic shaped-body of trehalose has free-flowing ability is a temperature of about 90°C to about 120°C or higher.

13. The process of claim 11 or 12, which uses said thermoplastic shaped-body of trehalose to said other substance in a ratio of not lower than 1.0 w/w %, on a dry solid basis.

14. The process of claim 11, 12 or 13, wherein said other substance is a material selected from the group consisting of food materials, cosmetic materials, pharmaceutical materials, plastic materials, and materials for agriculture, fishery, and live stocks.

15. The process of claim 11, 12, 13 or 14, wherein the method used in said forming step is a technique selected from the group consisting of stamping forming, cutting forming, crystallizing forming, injection molding, extrusion, blow

molding, plate forming, casting molding, pressure forming, compression molding, calendar molding, laminate molding, coating molding, rotational molding, spray molding, fixing molding, and blister molding.

16. The process of claim 11, 12, 14 or 15, wherein said shaped-body containing trehalose is used in food products, cosmetics, pharmaceuticals, agriculture, fishery, live stocks, gradually-disintegrable plastics, biodegradable shaped-bodies, or household articles.

17. A method for lowering the adhesion of a trehalose solution, comprising a step of, when concentrated by heating, either controlling the moisture content of said trehalose solution to a level not higher than about 8.3 w/w % in the presence of a substance capable of lowering the adhesion of said trehalose solution, or controlling the moisture content of said trehalose solution to a level not higher than about 2.4 w/w % in the absence of said substance.

18. The method of claim 17, wherein said substance capable of lowering the adhesion of said trehalose solution is a hydrophobic and/or hydrophilic organic substance.

19. The method of claim 17 or 18, wherein said organic substance is a member selected from the group consisting of lipids, carbohydrates, saccharides, polyalcohols, and emulsifiers.

20. The method of claim 17, 18 or 19, wherein said substance capable of lowering the adhesion of said trehalose solution is allowed to coexist in an amount of less than 40 w/w % to said trehalose, on a dry solid basis.

21. A thermoplastic shaped-body of trehalose, which

is obtainable by forming the trehalose solution whose adhesion has been lowered by the method of any one of claims 17 to 20.

22. The thermoplastic shaped-body of trehalose of claim 21, which has a free-flowing ability at temperatures of over 90°C as a temperature thereof.

[Detailed Description of the Invention]

[Field of the Invention]

The present invention relates to a thermoplastic shaped-body of trehalose, process and uses thereof, and more particularly, to a thermoplastic shaped-body of trehalose, process for producing the thermoplastic shaped-body of trehalose characterized in that it comprises a step of preparing the shaped-body of trehalose by forming a trehalose solution, process for producing a shaped-body containing trehalose comprising a step of heating the thermoplastic shaped-body of trehalose to impart it free-flowing ability and forming the resultant, method for lowering the adhesion of a trehalose solution, and uses thereof.

[Prior Art]

As disclosed in "*Chemical Industry*", pp. 57-65, June 1999, trehalose is being produced from starches as materials in an industrial scale and is widely used especially in food industries. However, methods for forming trehalose in use is restricted; Trehalose is merely used by mixing crystalline or powdery trehalose with fillers or binders and forming the resulting mixture into tablets; or dissolving by heating in water crystalline or powdery trehalose and other saccharide(s) in an amount exceeding trehalose, concentrating by boiling the resulting mixture, pouring the mixture into molds, cooling the

resultant to form candies. These methods require relatively-specific machines and relatively-high techniques; the obtained shaped-bodies are too monotonic in their shapes. As an arbitral-shape of product containing trehalose, for example, it is substantially difficult to produce a cotton-like shaped-body because commercially-available crystalline or powdery trehalose could not be processed with cotton-candy-machines and could not be processed well into cotton candies like those processed with rock candies. It has been demanded a process for producing shaped-bodies having arbitral shapes by using trehalose.

[Object of the Invention]

The object of the present invention provides a process for easily producing shaped-bodies having arbitral shapes by using trehalose.

[Means to Attain the Object]

To attain the above object the present inventors eagerly studied on the process for producing shaped-bodies of trehalose. As a result, they found that high-trehalose-content glassy shaped-bodies have higher storage stability than those processed with other saccharides, have desired thermoplasticity and satisfactory heat-resistance, have easiness of forming into shaped-bodies having arbitral shapes after heated to impart them free-flowing ability, and have easiness of forming repeatedly by using the above properties. Thus, the present inventors accomplished this invention. Based on the findings that high-trehalose-content glassy shaped-bodies have satisfactory thermoplasticity and stability, the present inventors solved the above object by providing a thermoplastic shaped-body of trehalose; a process for producing the thermoplastic shaped-body

of trehalose characterized in that it comprises a step of either forming a trehalose solution with a substance capable of lowering the adhesion of the trehalose solution and having a moisture content of not higher than about 8.3 w/w % (throughout the specification, the term "w/w %" is abbreviated as "%" hereinafter, unless specified otherwise), or forming a trehalose solution having no such a substance but a moisture content of not higher than about 2.4%; a process for producing a shaped-body containing trehalose by heating the above thermoplastic shaped-body of trehalose to impart it free-flowing ability and forming the resultant into a product having an appropriate shape along with or without other substance(s); a method for lowering the adhesion of a trehalose solution characterized in that it comprises a step of, when concentrated by heating, either lowering the moisture content of the trehalose solution to a level of not higher than about 8.3% in the presence of a substance capable of lowering the adhesion of the trehalose solution, or lowering the moisture content of the trehalose solution to a level of not higher than about 2.4%; and a thermoplastic shaped-body of trehalose obtainable by using these processes and methods.

[Preferred Embodiments of the Invention]

The trehalose used in the present invention is also called α,α-trehalose, and the origin is not restricted. For example, trehalose prepared from yeasts as disclosed in Japanese Patent Kokai No. 246,097/95, one prepared from maltose by using phosphorylases as disclosed in Japanese Patent Kokai No. 216,695/83, and ones prepared from starches by using enzymatic saccharifications as disclosed in Japanese Patent Kokai Nos.

170,977/95, 213,283/95, etc. Commercially available high-purity hydrous or anhydrous crystalline trehaloses, etc., can be arbitrarily used. For example, "TREHAOSE®", a high-purity hydrous crystalline trehalose commercialized by Hayashibara Shoji, Inc., Okayama, Japan, can be advantageously used.

The thermoplastic shaped-body of the present invention has satisfactory storage-stability at ambient temperature and moisture, does not substantially change on standing, and has properties of being easily formed into products with an appropriate shape after melted by heating to impart it free-flowing ability, as well as of maintaining its characteristics even when repeatedly shaped and melted twice or more and being easily regenerated.

The thermoplastic shaped-body of the present invention is a shaped-body which comprises a substance capable of lowering the adhesion of a trehalose solution and has a moisture content of not higher than about 8.3%, preferably, not higher than about 4.4%; or which comprises such a substance and has a moisture content of not higher than about 2.4%, preferably, not higher than about 2.0%. The above shaped-bodies become to be softened when heated to a temperature of about 70°C to about 90°C as a temperature thereof, and show free-flowing ability when heated to a temperature of about 90°C to about 120°C or higher. The process for producing the thermoplastic shaped-body of the present invention can be selected from the following appropriate ones: For example, the thermoplastic shaped-bodies can be prepared by providing hydrous or anhydrous crystalline trehalose; and directly or after moistened with appropriate amount of water, heating and melting or melting by heating the

resultant at a higher temperature as high as possible, preferably, at a temperature over 160°C, more preferably, a temperature of about 165°C to about 230°C, most preferably, a temperature of about 170°C to 220°C, to make into a glassy state. In addition, the shaped bodies can be prepared by providing aqueous trehalose solutions prepared by the above enzymatic reactions, those prepared by hydrogenating the aqueous trehalose solutions, those prepared by dissolving hydrous crystalline trehalose in water; preferably, those having a moisture content of at least 30%, more preferably, a moisture content of 30% but below 50%, corresponding to those with a trehalose concentration of below 70%, more preferably, about 50% but below 70%; concentrating by heating the resulting trehalose solutions under normal or reduced pressure into high-concentrated trehalose solutions; and allowing the solutions to make into their glassy states.

In the case of concentrating the aqueous trehalose solutions by heating, the moisture content and the solution temperature are closely related each other; Under normal pressure, the temperatures of the aqueous trehalose solutions are 115°C, 125°C, 145°C, 165°C and 185°C which correspond to the moisture contents of 15.3%, 9.9%, 5.2%, 2.4% and 1.1%, respectively. During the process for examining their relationship, the present inventors found an unexpected phenomenon that the adhesion of aqueous trehalose solutions greatly changed depending on the moisture content of or the temperature of the solutions when concentrated by heating. The findings are: The adhesion of trehalose solutions increases as the moisture content of the solutions decreases to a level

below about 10% when concentrated by heating, particularly, highly increases when the moisture content decreases to about 8.3% or lower, then reaches to the maximum level when the moisture content becomes to about 4.4% to about 3.0% after further concentrated and the free-flowing ability of the solution decreases. When the trehalose solutions are further continuously concentrated by heating, the adhesion of the solutions greatly decreases when the moisture content becomes to less than 3%, and the adhesion and the free-flowing ability of the solutions are improved to an extent that does not hinder the handleability of their forming when the moisture content becomes to about 2.4% or lower, preferably, to about 2.0% or lower.

To improve the handleability by lowering the extraordinary-increased adhesion of trehalose solutions, the present inventors energetically studied and found that the coexistence of a substance capable of lowering the adhesion of trehalose solutions is advantageous, and this effectively decreases the adhesion of high-purity trehalose solutions by a large margin and lowers the foaming of the solutions during their concentration by heating, resulting in improvement of handleability and easiness of the process for preparing shaped-bodies.

The substances capable of lowering the adhesion of the above trehalose solutions usable in the present invention include any substances as long as they lower the adhesion of the trehalose solutions. For example, preferable substances are organic substances selected from hydrophobic substances such as lipids and carbohydrates, hydrophilic substances such as

saccharides and polyalcohols, and emulsifiers having both hydrophobic- and hydrophilic-properties.

The lipids used in the present invention can be preferably one or more members from oils, fats and fatty acids; hydrocarbons, which are in liquid forms at about 100°C to 230°C, such as alkanes, alkenes and heterocyclic compounds; saccharides such as monosaccharides and oligosaccharides excluding trehalose, mono- and di-saccharide alcohols; polyalcohols such as glycerine, diethylene glycol, and propylene glycol; and emulsifiers such as sucrose fatty acid esters, glycerine fatty acid esters, poly glycerine fatty acid esters, and lecithin.

As described above, the thermoplastic shaped-body of trehalose of the present invention can be produced by either concentrating trehalose solutions to give a prescribed moisture content, preferably, to a moisture content of not higher than about 8.3%, by using the method for lowering the adhesion of the solutions by coexisting the above substances capable of lowering the adhesion of the solutions, and forming the concentrates into shaped-bodies in a glassy state; or concentrating trehalose solutions to give a preferable moisture content of not higher than about 2.4% without coexisting the substance capable of lowering the adhesion of the solutions and forming the concentrates into shaped-bodies in a glassy state. The method of forming used arbitrarily in the present invention includes, for example, conventional ones such as stamping molding, cutting molding, atmospheric molding, molding in water or oils, extrusion, pressure forming, particularly, biaxial extrusion, etc.

To keep satisfactory shelf-life and thermoplasticity,

the thermoplastic shaped-body thus obtained should preferably contain trehalose as much as possible; usually, those which contain trehalose in an amount over 60%, d.s.b., preferably, not less than about 70%, more preferably, not less than about 80%, and more preferably, not less than about 90%.

The form of the thermoplastic shaped-body used preferably in the present invention can be those which are easily processed on conventional forming- or molding-machines or apparatuses when the thermoplastic shaped-body as a material is heated to show free-flowing ability and formed into an appropriate shape; and those which can be easily heated, and if necessary, after pressed or reduced, to easily melt to show free-flowing ability, and formed to impart it an appropriate shape by using the above machines or apparatuses.

Preferable concrete examples of the forms of the thermoplastic shaped-body are usually those having a short diameter of about 0.1-20 mm, and preferably, about 2-15 mm, and a longer diameter of equal to or longer than the short diameter. As the forms of the thermoplastic shaped-body, granules, spheres, hemispheres, elliptical spheres, short rods, short pyramids, half corns, cubes, rectangular parallelepipeds, short tubes, and fibers can be arbitrarily used. Generally, the form called pellet is preferably used.

The thermoplastic shaped-body thus obtained generally contains a substance capable of lowering the adhesion of trehalose solutions and has a moisture content of not higher than about 8.3%, preferably, not higher than about 4.4%; or has a moisture content of not higher than about 2.4%, preferably, not higher than about 2.0% without containing the above

substance. In either case, the thermoplastic shaped-body contains trehalose in an amount of over 60%, d.s.b., and preferably, in an amount of not lower than about 70%, d.s.b., wherein the trehalose is amorphous and transparent or translucent; relatively-high in moisture-resistant, heat-resistant, acid-resistant, alkaline-resistant, and shelf-life; and easily handleable. The thermoplastic shaped-body of the present invention can be used, for example, by dissolving in a small amount of hot water into a high trehalose content paste, and further used as a base for pulverization by coexisting a trehalose crystal seed in the resulting mixture to crystalize trehalose to facilitate the pulverization.

The most valuable use of the thermoplastic shaped-body of the present invention is for the use using its thermoplasticity; the thermoplastic shaped-body can be arbitrarily reheated to melt to impart it free-flowing ability, then directly formed into appropriate shapes; or can be arbitrarily formed into shaped-bodies containing trehalose by processing with other substances in such a manner of coating, mixing, fixing, inserting, enclosing, etc.

Varying depending on the kind or content of the substances capable of lowering the adhesion of trehalose solutions, the heating conditions used in the present invention are those which melt and soften the thermoplastic shaped-bodies of trehalose, preferably, which make them into their liquid states or free-flowing conditions for ease of forming into appropriate shapes. The softening temperature is generally adjusted by heating to about 70°C to about 90°C for a product temperature, while the temperature for imparting free-flowing

ability is adjusted by heating to about 90°C or higher, and preferably about 100°C to about 120°C or higher for a product temperature. In general, when the thermoplastic shaped-bodies have a relatively-high moisture content of not higher than about 8.3% but over 2.4%, the temperature for imparting free-flowing ability is set to a relatively-low temperature, generally, to a temperature of at least about 90°C, and preferably to a temperature of about 100°C to about 160°C. While in the case of having a relatively-low moisture content of about 2.4% or lower, the thermoplastic shaped-bodies are heated to a relatively-high temperature, generally, to a temperature of about 100°C or higher, and preferably to a temperature selected from about 110°C to about 230°C.

When formed along with other substances, the thermoplastic shaped-bodies of trehalose can be selectively treated, for example, by first mixing with other substances, then heating to impart them free-flowing ability and forming into appropriate shapes; or first heating only the thermoplastic shaped-bodies of trehalose to impart them free-flowing ability, then mixing with other substances and forming into appropriate shapes. If necessary, the thermoplastic shaped-bodies of trehalose can be moistened or admixed with a small amount of water to lower their melting temperatures or to accelerate the mixing.

Any substances can be arbitrarily used as the other substances used in the present invention as long as they can be formed into stable shaped-bodies of the present invention when used in combination with the thermoplastic shaped-bodies of trehalose; materials and crude ingredients including additives

generally used in the fields including chemical industries where the thermoplastic shaped-bodies of trehalose can be used, for example, in the fields of food products, cosmetics, pharmaceuticals, agriculture, fishery, live stocks, plastics, household articles, etc. More particularly, the followings, which are generally used in one or more of the aforesaid fields, can be selectively used depending on purposes: For example, a sweetener, coloring agent, color-developing agent, fermentation-controlling agent, flavor, aromatic, enhancer, baking powder, preservative, sterilizer, oxidation-preventing agent, antioxidant, bleaching agent, gum, stabilizer, emulsifier, tenderizer, quality-improving agent, seasoning, spice, wax, bitter agent, sour agent, processing aid, enzyme preparation, gum base, surfactant, plasticizer, smooth and gloss-imparting agent, solubilizing agent, reductant, buffer, base, adsorbent, taste-imparting agent, binder, suspending agent, coating agent, lubricant, lubricant-controlling agent, filler, anti-foaming agent, refreshing agent, adhesive, enhancer, mastication agent, sugar-coating agent, isotonicity-imparting agent, sticking agent, stickiness enhancer, agent for consistency, consistency-imparting agent, inflammatory inhibitor, heat-generating agent, foaming agent, skin protective, excipient, disperser, pH-controlling agent, decaying agent, decaying aid, anti-rust agent, anti-humectant, antiseptic, fungicidal, preservative, pain-removing agent, vitamin, antibiotic, chemotherapeutic agent, anti-allergic, anti-histaminic, antalgic, antipruritic, astringent, anti-inflammatory, steroid, hormone, cardiac, dental/oral agent, anastaltic, ultraviolet absorber, algicide, thermoplastic resin, synthetic resin, crystallization-preventing

agent, deterioration-preventing agent, retrogradation-preventing agent, burning preventive, resin-hardening agent, coagulant, foaming agent, foaming stabilizer, anti-static agent, curing agent, etc. In particular, examples of the above materials and crude ingredients used in the field of food products are saccharides such as starches, amyloses, amylopectins, pregelatinized starches, dextrins, substantially-indigestive dextrins, starch derivatives, pullulan, pectin, alginic acid, polysaccharides, dietary fibers, monosaccharides, oligosaccharides including functional oligosaccharides, and sugar alcohols; proteins, peptides, amino acids, oils and fats, and minerals.

The more the trehalose content in the thermoplastic shaped-bodies of trehalose obtained by forming with other substances, the more they are preferably used since trehalose present in the shaped-bodies generally improves the stability of relatively-unstable functional substances and effective ingredients such as the aforesaid vitamins, lipids, proteins, effective substances, and physiologically-active substances. In general, the trehalose content is relatively-low in the shaped-bodies which the surfaces are coated with trehalose, while the content is relatively-high in those prepared by homogeneously mixing trehalose with other substances and solidifying the whole contents. In the present invention, since the thermoplastic shaped-bodies of trehalose should be formed or molded into appropriate shapes, they are generally incorporated into other substances in an amount of at least about 1.0%, preferably, at least about 2.0%, more preferably, at least about 5.0%, and more preferably, at least about 10% to

the other substances, d.s.b. The form of the resulting shaped-bodies is appropriately selected from those which comprise point-, line-, face-, and stereoscopic-structures such as granules, rods, plates, sheets, films, tubes, laminates, and sponges.

Depending on final shapes, the method for forming the above shaped-bodies is appropriately selected from conventional ones such as stamping molding, cutting molding, injection molding, extrusion, blowing molding, plate molding, casting molding, compression molding, pressure forming, calendar molding, laminating molding, film forming, rotation molding, spraying molding, fixing molding, blister molding, etc.

Depending on other substances used in combination and functional substances and effective ingredients contained therein, the above shaped-bodies containing trehalose obtained by transforming into other forms can be arbitrarily used after formed into food products, cosmetics, pharmaceuticals, products for agriculture, fishery, and live stocks, gradually-disintegrable plastics, biodegradable shaped-bodies, and house articles, etc.

The following experiments describe the present invention:

Experiment 1

Influence of boiling down temperature or moisture content of saccharide solutions on their adhesiveness and storage stability of shaped bodies formed therewith

As saccharides, non-reducing saccharides, i.e., anhydrous crystalline maltitol and crystalline trehalose dihydrate were provided, respectively placed in a pan, and

heated after admixed with water into a 50% aqueous saccharide solution. The aqueous solutions were continued heating to boil and concentrate. When the saccharide solutions reached to respective temperatures of 120°C, 130°C, 140°C, 150°C, 160°C, 170°C, 180°C and 190°C, a glass rod having a diameter of 4 mm was inserted into each saccharide solution three centimeters in depth from the liquid surface, followed by holding the glass rod thereat for three seconds and then pulling up gently. The weight of the saccharide solution adhered to the glass rod was calculated by weighing the total weight of the glass rod and the saccharide solution and reducing only the weight of the glass rod from the total weight. Each saccharide solution boiled down was sampled and placed in a small depositor and cooled to ambient temperature into a shaped-body as a sample, 20 mm in length, 15 mm in width, and 5 mm in thickness. Shaped-bodies formed within 24 hours after processing were measured for moisture. The moisture content was determined by the diatomaceous earth method in a usual manner.

A portion of each of the same samples was placed in a Petri dish and allowed to stand at a relative humidity of about 70% and at 25°C for 10 days before observing the presence of adhesion.

The level of adhesion was evaluated by examining the level of the samples adhered to the Petri dishes after absorbing moisture and classifying the levels into two ranks of "Yes" and "No" as a rough indication of storage stability. The results are in Table 1.

Table 1

Saccharide	Item measured	Boiling down temperature (°C under normal pressure)									
		120	130	140	150	155	160	165	170	180	190
Trehalose	Adhered content (g)	0.230	0.569	0.890	1.250	1.474	1.726	1.580	0.915	0.761	0.579
	Moisture content (%)	12.1	8.3	6.1	4.4	3.6	3.0	2.4	2.0	1.3	0.9
	Adhesion	No	No	No	No	No	No	No	No	No	No
Maltitol	Adhered content (g)	0.200	0.317	0.338	0.369	0.355	0.335	0.345	0.313	0.294	0.257
	Moisture content (%)	9.1	5.5	4.1	3.2	2.7	2.5	2.1	1.9	1.4	0.9
	Adhesion	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes

As evident from Table 1, it was found that, in the case of trehalose, the weight of trehalose solution adhered to the glass rod unexpectedly and remarkably increased from 120°C, corresponding to a trehalose solution with a moisture contents of about 12%, to 160°C, corresponding to a trehalose solution with a moisture contents of about 3.0%, respectively; while the moisture content decreased at temperatures over the above temperature range, meaning that the trehalose solution less adhered by a large margin to the glass rod as the temperature increased from 160°C to 190°C in spite of the increment of trehaose concentration. It was found that the adhesive of trehalose solution reached to a maximum level at temperatures from about 150°C to about 160°C, corresponding to a trehalose solution with a moisture content of about 4% to about 3%, then gradually decreased at temperatures of over 160°C, corresponding to a trehalose solution with a moisture contents of less than about 3%.

In the case of the maltitol solution as a control, it was observed that the amount of the maltitol solution adhered to the glass rod slightly increased at temperatures from 120°C to 150°C and decreased at temperatures from 150°C to 190°C, and found that the level of alteration was negligible as compared with trehalose solution. Based on these data, it was revealed that, when trehalose solution is boiled down, the adhesion of trehalose solution highly increases up to 160°C and this hinders the processibility during its forming or molding, and that the drawback should be improved. It was also found that the adhesion of trehalose solution decreased when boiled down at temperatures of 165°C or higher, corresponding to a trehalose

solution with a moisture content of about 2.4% or lower, and the free-flowing ability was remarkably improved to ease the processibility of the handling of trehalose solution by a large margin. It was found that, unlike shaped-bodies processed with maltitol solution, those with trehalose solution do not substantially change on standing when stored at a relative humidity of 70% and at ambient temperature and have satisfactory moisture-resistant and storage stability.

Experiment 2

Method for lowering adhesion

Five hundred and fifty parts by weight of crystalline trehalose dihydrate were placed in a stainless-steel pan, admixed with, as a substance capable of lowering adhesion of trehalose solution, 50 parts by weight of safflower oil, 50 parts by weight of salad oil, 150 parts by weight of anhydrous crystalline maltitol, or 158 parts by weight of crystalline lactitol monohydrate, and further mixed with 450 parts by weight of water, followed by completely dissolving the saccharides while heating. Then, the heating was continued to boil down each saccharide solution. When reached to temperatures of 120°C, 130°C, 140°C, 150°C, 160°C, 170°C and 180°C, the saccharide solutions were subjected to measurement of the weight of each saccharide solution adhered to a glass rod similarly as the method in Experiment 1. The results are in Table 2 where the trehalose content in each saccharide solution is shown in parallel, based on a dry solid.

Table 2

Saccharide	Coexisting substance (Trehalose content (%, d.s.b.)	Item measured	Boiled down temperature (°C under normal pressure)						
			120	130	140	150	160	170	180
Trehalose	Non (100%)	Adhered content (g)	0.230	0.569	0.890	1.250	1.726	0.915	0.761
	Safflower oil (about 91%)	Adhered content (g)	0.150	0.194	0.225	0.256	0.257	0.277	0.278
	Salad oil (about 91%)	Adhered content (g)	0.195	0.499	0.512	0.596	0.538	0.481	0.428
	Maltitol (about 77%)	Adhered content (g)	0.230	0.538	0.861	0.987	0.786	0.607	0.570
	Lactitol (about 77%)	Adhered content (g)	0.265	0.466	0.773	0.862	0.919	0.893	0.574

As evident from Table 2, it was found that the coexistence of any of safflower oil, salad oil, maltitol, and lactitol significantly inhibited the remarkable increment of the adhesion of trehalose solution at temperatures from 120°C to 160°C, particularly, temperatures from 130°C, corresponding to a trehalose solution with a moisture content of about 8.3%, to 160°C, corresponding to a trehalose solution with a moisture content of about 3.0%; and strongly lowered the adhesion. Particularly, it was found that the adhesion peaks of all the trehalose solutions at temperatures in the range of about 150°C, corresponding to a trehalose solution with a moisture content of about 4.4%, to about 160°C, corresponding to a trehalose solution with a moisture content of about 3.0%, remarkably lowered, resulting in strong reduction of their adhesion.

It was also found that, under the coexistence of the above substances, the adhesion of all the trehalose solutions at temperatures of over 160°C was lower than that of the trehalose solutions alone.

Based on these, it was found that the strong increment of adhesion of trehalose solutions observed during their boiling down at temperatures from 130°C, corresponding to a trehalose solution with a moisture content of about 8.3%, to 160°C, corresponding to a trehalose solution with a moisture content of about 3.0%, is remarkably inhibited by coexisting substances capable of lowering the adhesion of trehalose solution, for example, oils and fats such as safflower oil and salad oil, and saccharides such as maltitol and lactitol. Particularly, the adhesion peak of trehalose solution observed at temperatures from about 150°C, corresponding to a solution with a moisture

content of about 4.4%, to about 160°C, corresponding to a trehalose solution with a moisture content of about 3.0%, remarkably lowered; and more particularly, even under higher temperatures of over 160°C for boiling down, the substances more reduced the adhesion of trehalose solution and remarkably improved the free-flowing ability, resulting in a conclusion that the substances outstandingly improved the handleability of thermoplastic shaped-bodies of trehalose during the processings.

The process for producing the thermoplastic shaped-bodies of trehalose according to the present invention, and some examples of uses thereof are disclosed in the below:

Example 1

Short-rod shaped-body

One hundred and ten parts by weight of "TREHAOSE®", a high-purity crystalline trehalose hydrate commercialized by Hayashibara Shoji, Inc., Okayama, Japan were placed in a dissolving tank, and admixed with and dissolved by heating in 90 parts by weight of water. The solution was concentrated by heating into a concentrate with a temperature of about 190°C and a moisture content of about 0.9% to lower the adhesion of the concentrate. Then, the resulting concentrate was formed by feeding to an extruder, cooled to ambient temperature to obtain a short-rod thermoplastic shaped-body of trehalose, about 2 mm in diameter and about 4 mm in length. Since the product was a colorless, transparent or translucent, and substantially amorphous shaped-body having satisfactory thermoplasticity and storage-stability under indoor circumstances, it can be arbitrarily reheated to a temperature of about 90°C or higher to impart it free-flowing ability, then transformed or processed

into a shaped-body with an appropriate form with or without using other substances having different functions. The product can be also used arbitrarily for preparing a paste containing trehalose by dissolving in a relatively-small amount of hot water, and the resulting paste can be used intact in an amorphous form or used as a powdered base after crystallization of trehalose.

Example 2

Elliptic spherical shaped-body

Similarly as in Example 1, a high-purity crystalline trehalose hydrate was dissolved by heating, and the solution was concentrated by heating, admixed to lower the adhesion of the solution with an olive oil in an amount of about 5% to trehalose, d.s.b., when reaching roughly to a temperature over 110°C at which the adhesion of the concentrate began to increase, and continued concentrating by heating until reaching to a temperature of about 170°C, corresponding to a moisture content of about 2.0%. The resulting concentrate was fed to a stamping-forming machine to obtain an elliptic-spherical thermoplastic shaped-body of trehalose, about 15 mm in length. Since the product was a colorless, transparent or translucent, and substantially amorphous shaped-body having thermoplasticity and satisfactory storage-stability under indoor circumstances, it can be arbitrarily heated to a temperature of about 90°C or higher to impart it free-flowing ability, then transformed or processed into a shaped-body with an appropriate form with or without using other substances having different functions. The product can be also used arbitrarily for preparing a paste containing trehalose by dissolving in a relatively-small amount

of hot water, and the resulting paste can be used intact in an amorphous form or used as a powdered base after crystallization of trehalose.

Example 3

Short square-pillar shaped-body

Similarly as in Example 1, a high-purity crystalline trehalose hydrate was dissolved by heating, and the solution was concentrated by heating, admixed to lower the adhesion of the solution with propylene glycol in an amount of about 3% to trehalose, d.s.b., when reaching roughly to a temperature of over 110°C at which the adhesion of the concentrate began to increase, and continued heating to boil down until reaching to a temperature of about 165°C, corresponding to a moisture content of about 2.4%. The resulting concentrate was fed to a cutting-forming machine to obtain a short square-pillar thermoplastic shaped-body, a side length of about 5 mm and about 10 mm in height. Since the product was a colorless, transparent or translucent, and substantially amorphous shaped-body having thermoplasticity and satisfactory storage-stability under indoor circumstances, it can be arbitrarily reheated to a temperature of about 90°C or higher to impart it free-flowing ability, then transformed or processed into a shaped-body with an appropriate form with or without using other substances having different functions. The product can be also used arbitrarily for preparing a paste containing trehalose by dissolving in a relatively-small amount of hot water, and the resulting paste can be used intact in an amorphous form or used as a powdered base after crystallization of trehalose.

Example 4

Hemispheric shaped-body

According to the method disclosed in Japanese Patent Kokai No. 73,482/96, a seed culture of a mutant of *Rhizobium* sp. M-11 strain, FERM BP-4130, was inoculated to and cultured in a nutrient culture medium for about 70 hours by a fermenter. After culturing, the resulting culture was filtered to remove cells using an SF membrane to yield an about 100ℓ culture supernatant, followed by concentrating the supernatant with a UF membrane to obtain an about five liters of a crude enzyme concentrate containing about 410 units/ml of a non-reducing saccharide-forming enzyme and about 490 units/ml of a trehalose-releasing enzyme. Corn starch was suspended into an about 33% starch suspension which was then subjected to the action of α-amylase to obtain a liquefied solution with a DE (dextrose equivalent) of about four. To the liquefied solution were added 0.02 ml/g starch of the above crude enzyme concentrate, 500 units/g starch of isoamylase, and five units/g starch of cyclomaltodextrin glucanotransferase, then subjected to an enzymatic reaction at pH 6.2 and at 40°C for 48 hours. The reaction mixture was heated to inactivate the remaining enzymes, then admixed with 10 units/g substrate of glucoamylase, and enzymatically reacted at 50°C for 10 hours. The resulting reaction mixture, containing 85.6% trehalose, d.s.b., was heated to inactivate the remaining enzyme, and according to conventional manner, purified by decoloration and desalting, and concentrated into a 45% syrup, i.e., a relatively-low reducing saccharide syrup enriched with trehalose. According to conventional manner, the syrup was hydrogenated, purified by decoloration and desalting, and concentrated into a syrup

containing about 85% trehalose, d.s.b., and others mainly composed of sorbitol. The syrup was concentrated and boiled down by heating to a temperature of about 180°C or a moisture content of about 1.3% and to lower its adhesion, and the concentrate was dropped into salad oil through a fine tube and solidified by cooling to obtain a hemispheric thermoplastic shaped-body of trehalose, about 8 mm in base diameter. Since the product was a colorless, transparent or translucent, and substantially amorphous shaped-body having thermoplasticity and satisfactory storage-stability under indoor circumstances, it can be arbitrarily reheated to a temperature of about 90°C or higher to impart it free-flowing ability, then transformed or processed into a shaped-body with an appropriate form with or without using other substances having different functions. The product can be also used arbitrarily for preparing a paste containing trehalose by dissolving in a relatively-small amount of hot water, and the resulting paste can be used intact in an amorphous form or used as a powdered base after crystallization of trehalose.

Example 5

Fibrous shaped-body

Using an electric furnace, crystalline trehalose hydrate was melted by heating to a temperature of about 200-210°C or to a moisture content of about 0.5% or lower. While pouring out of an outlet provided in the basement of the furnace, the melt was blown by air to obtain a fibrous shaped-body, about 0.1-0.2 mm in diameter, followed by disrupting the fibrous shaped-body into a short-rod fibrous shaped-body, less than about 20 mm in length. Since the product was a colorless,

transparent or translucent, and substantially amorphous shaped-body having thermoplasticity and satisfactory storage-stability under indoor circumstances, it can be arbitrarily reheated to a temperature of about 100°C or higher to impart it free-flowing ability, then transformed or processed into a shaped-body with an appropriate form with or without using other substances having different functions. The product can be also used arbitrarily for preparing a paste containing trehalose by dissolving in a relatively-small amount of hot water, and the resulting paste can be used intact in an amorphous form or used as a powdered base after crystallization of trehalose.

Example 6

Short-rod shaped-body

A thermoplastic shaped-body of trehalose obtained by the method in Example 3 was heated to about 120°C and fed to an extruder to obtain a short-rod thermoplastic shaped-body of trehalose, about 2 mm in diameter and about 4 mm in length. Since the product was a colorless, transparent or translucent, and substantially amorphous shaped-body having a satisfactory thermoplasticity and storage-stability under indoor circumstances, it can be arbitrarily reheated to a temperature of about 90°C or higher to impart it free-flowing ability, then transformed or processed into a shaped-body with an appropriate form with or without using other substances having different functions. The product can be also used arbitrarily for preparing a paste containing trehalose by dissolving in a relatively-small amount of hot water, and the resulting paste can be used intact in an amorphous form or used as a powdered base after crystallization of trehalose.

Example 7

Cotton-like shaped-body

To one part by weight of a thermoplastic shaped-body of trehalose, obtained by the method in Example 2, was adhered by homogeneously spraying 0.05 part by weight of a concentrated lemon-juice, dried, and fed to a cotton-candy machine and heated to melt at temperatures over about 100°C to obtain a cotton candy. Unlike conventional cotton-candy processed with sugar, the product is an acid-punched, lemon-flavored, cotton-candy containing trehalose.

Example 8

Coated shaped-body

One hundred parts by weight of wheat flour, two parts by weight of yeast, five parts by weight of sugar, one part by weight of maltose, and 0.1 part by weight of yeast food were in a usual manner kneaded with water. The mixture was fermented at 26°C for two hours, then further aged for 30 min, followed by rounding with hands the resulting dough for bread. A thermoplastic shaped-body of trehalose, obtained by the method in Example 4, was partially inserted into the dough from its surface in an amount of about 2% to the dough, d.s.b. According to conventional manner, the dough was baked at about 180°C. The product is a unique and interesting bread having a melt-distorted shaped-body of trehalose on its surface, and is a high-quality bread having a satisfactory appearance and texture, as well as an adequate elasticity and mild sweetness.

Example 9

Coated shaped-body

Sixty parts by weight of butter, 20 parts by weight

of sugar, 20 parts by weight of maltose, and 10 parts by weight of a crystalline trehalose dihydrate powder were well kneaded, then mixed with 40 parts by weight of chicken egg to obtain a creamy product. To the product were added 140 parts by weight of soft flour, 10 parts by weight of corn starch, and three parts by weight of pullulan. The resulting dough for cookie was passed through a mum cap over a plain plate to form a flower-shaped product, followed by adhering over the product a thermoplastic shaped-body of trehalose, obtained by the method in Example 5, in an amount of about one percent, d.s.b., to the dough, and baking the resulting product at about 170°C for 15 min. In the baked product having a satisfactory flavor, taste and mouth feel, the cookie dough and the unique melt-distorted shaped-body containing trehalose well adhered each other without separation.

Example 10

Granular shaped-body

A mixture powder, consisting of 15 parts by weight of a thermoplastic shaped-body of trehalose obtained by the method in Example 2, two parts by weight of plain yogurt, and 10 parts by weight of anhydrous trehalose powder, was heated to about 100°C and fed to a biaxial extruder to produce a granular shaped-body containing trehalose. The product is a yogurt-flavored sweet shaped-body and can be arbitrarily used as a confectionery material for premixes, frozen deserts, cakes, etc., and as a therapeutic nutrient for intubation feedings; and can be incorporated into whipped margarine creams, spreads, cheese cakes, jellies, etc. to obtain a yogurt-flavored food product.

Example 12

Elliptic-spherical shaped-body

A mixture, consisting of 500 parts by weight of a thermoplastic shaped-body of trehalose obtained by the method in Example 2, 270 parts by weight of an egg york powder, 209 parts by weight of a skim milk powder, 4.4 parts by weight of sodium chloride, 1.85 parts by weight of potassium chloride, four parts by weight of magnesium sulfate, 0.01 part by weight of thiamine, 0.1 part by weight of sodium ascorbate, 0.6 part by weight of vitamin E acetate, and 0.04 part by weight of nicotinic acid amide, were heated to about 100°C and fed to a pressure-forming machine to obtain an elliptic-spherical shaped-body containing trehalose. The product, weighing about five grams, is satisfactorily used as a nutrient shaped-body by melting in the mouth as a therapeutic nutrient for sick persons and as a nutrient for healthy persons to maintain and promote their heath.

Example 13

Granular shaped-body

A mixture of 10 parts by weight of a thermoplastic shaped-body of trehalose, obtained by the method in Example 1, and one part by weight of a sodium carbonate powder was heated to about 100°C and fed to a biaxial extruder to produce a granular shaped-body containing trehalose. The product can be widely used in a variety of uses as an easily handleable pH-controlling agent with less fear of causing scattering of the sodium carbonate powder.

Example 14

Short-tubular shaped-body

A mixture of 100 parts by weight of a thermoplastic shaped-body of trehalose obtained by the method in Example 2, 200 parts by weight of fish flour, 20 parts by weight of pregelatinized starch, three parts by weight of pullulan, and 10 parts by weight of water was heated to about 100°C and fed to an extruder to obtain a short-tubular shaped-body containing trehalose. The product was soaked in a solution of about 10% shellac in alcohol, then instantly taken out from the solution, and dried with hot air to obtain a short-tubular shaped-body containing trehalose. The product is a feed with lesser fear of spoiling water and can be satisfactorily used as a feed for breeding or enjoying fishes, shellfishes, and cruschymata, and crustaceans independently of freshwater or sea water.

Example 15

Flowerpot

A mixture of 100 parts by weight of a thermoplastic shaped-body of trehalose obtained by the method in Example 6, 15 parts by weight of glycerine, and 50 parts by weight of used-paper pulp was heated to about 150°C and fed to an injection molder to obtain a flowerpot. The resulting flowerpot was soaked in a melted wax and cooled to produce a flowerpot containing trehalose. Since the product is gradually degradable and biodegradable, it can be advantageously used as a flowerpot for transplanting plants therewith without damaging the plants' roots.

Example 16

Short-rod shaped-body

A mixture of 60 parts by weight of a thermoplastic shaped-body of trehalose, obtained by the method in Example 1,

30 parts by weight of a coral powder, and two parts by weight of corn steep liquor was heated to about 110°C and fed to an extruder to obtain a plant activator of short-rod shaped-body containing trehalose. The product is satisfactorily used as a durable plant-activator because it needs no container, has easy handleability, and in use dissolves and disintegrates in an appropriate rate.

Example 17

Golf tee

A mixture of 10 parts by weight of a thermoplastic shaped-body of trehalose, obtained by the method in Example 4, and 0.5 part by weight of pullulan was heated to about 160°C and fed to an injection molder to form a golf tee which was then soaked in a solution of about 10% shellac in alcohol and dried to obtain the captioned product. After crashed into pieces by shot, the product is gradually disintegrated by rainwater and biodegraded. Thus, the product does not spoil the beauty and environment of golf course. In addition, adequately dispersed trehalose contained in the golf tee activates the growth of lawn in the golf course.

Example 18

Film shaped-body

A mixture of 92 parts by weight of low-density polyethylene, eight parts by weight of a thermoplastic shaped-body of trehalose obtained by the method in Example 3, 0.05 part by weight of calcium stearate, 0.5 part by weight of an enzyme-treated rutin, and 0.5 part by weight of a tea extract was heated to about 120°C and fed to a pressure forming machine to obtain a short-rod shaped-body. Thereafter, the shaped-body was

heated to about 130°C and fed to an extruder as inflation technique to obtain a film shaped-body. The product can be arbitrarily used as a film for keeping the freshness of fresh plants and foods such as flowers, vegetables, fruits, meets, and fishes.

[Effect of the Invention]

As described above, the thermoplastic shaped-body of trehalose according to the present invention is easily handleable because of its satisfactory storage-stability, thermoplasticity, and heat resistance. Using the thermoplasticity, the product has a characteristic of being repeatedly formed or molded.

In producing the thermoplastic shaped-body of trehalose, the processibility during its forming or molding can be advantageously improved by lowering the adhesion of trehalose solution.

By heating the thermoplastic shaped-body of trehalose to impart it free-flowing ability, it can be easily formed into shaped-bodies with appropriate shapes. In the case of forming, the thermoplastic shaped-body of trehalose can be arbitrarily formed after mixing with functional substances and effective ingredients such as materials for food products, cosmetics, pharmaceuticals, agriculture, fishery, live stocks, and plastic-moldings.

The shaped-bodies thus obtained can be easily formed into products with appropriate shapes, improved their supplemental values because trehalose stabilizes relatively-unstable functional substances and effective ingredients. In addition, the shaped-bodies can be processed into gradually-

disintegrable, biodegradable ones, meaning that they are the products that well harmonize with the earth.

Accordingly, the present invention greatly influences on the fields of food-, cosmetic-, pharmaceutical-, agriculture-, fishery-, live stock-, plastic-, and house hold-industries.

[Document Name] Abstract

[Summary]

[Object] The object of the present invention is to easily form a shaped-body with an appropriate shape.

[Construction] The present invention solves the above object by providing a thermoplastic shaped-body of trehalose; a process for producing the thermoplastic shaped-body using a trehalose solution; a process for producing a shaped-body containing trehalose characterized in that it comprises the steps of heating the thermoplastic shaped-body of trehalose to impart it free-flowing ability, and forming the resultant into a shaped-body with an appropriate form; a method for lowering the adhesion of trehalose solution; and uses thereof.

[Selected Figure] None